Development of an accelerator source of epithermal neutrons for boron neutron capture therapy

S. Yu. Taskaev[1]

Budker Institute of Nuclear Physics, pr. Lavrentieva 11, Novosibirsk, 630090 Russia

Novosibirsk State University, ul. Pirogova 2, Novosibirsk, 630090 Russia

Представлен обзор современного состояния развития ускорительных источников эпитепловых нейтронов для бор-нейтронозахватной терапии (БНЗТ) – перспективной методики лечения злокачественных опухолей. Особое внимание уделено источнику эпитепловых нейтронов на основе нового типа ускорителя заряженных частиц – ускорителя-тандема с вакуумной изоляцией, и литиевой нейтроногенерирующей мишени.

We review the current status of the development of accelerator sources of epithermal neutrons for boron neutron capture therapy (BNCT), a promising method of malignant tumor treatment. Particular attention is given to the source of epithermal neutrons based on a new type of charged particle accelerator: tandem accelerator with vacuum insulation and lithium neutron-producing target.

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INTRODUCTION

According to the World Health Organization, cancer incidence has been steadily increasing and leads to significant mortality. Drug development and treatment of malignant tumors is an important and hitherto not solved scientific problem. Boron neutron capture therapy (BNCT) is a promising approach in the treatment of a variety of malignant tumors, especially intractable brain tumors, exerting extremely effective selective impact directly on cancer cells [1, 2].

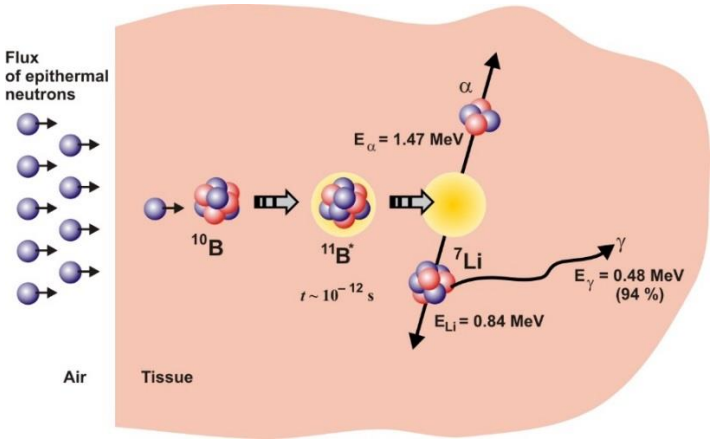

Fig. 1. Schematic representation of the BNCT principle

BNCT is a binary form of radiation therapy that uses a unique high ability of nonradioactive boron-10 nucleus to absorb thermal neutrons. The cross section of this absorption reaction is 3837 b. The absorption of the neutron by the ^{10}B nucleus leads to an instant nuclear reaction ^{10}B(n,α)^{7}Li with an energy release of 2.79 MeV. In 6.1% cases, the energy is distributed only between the nuclei of lithium and α-particles, in 93.9% of cases the lithium nucleus is emitted in an excited state and emits a γ-quantum with an energy of 0.48 MeV (Fig. 1). The products of the nuclear reaction, namely the nucleus of lithium with an energy of 0.84 MeV and α-particle with an energy of 1.47 MeV, are characterized by high stopping power (averages 162 and 196 keV μm^{-1}, respectively) and a small

[1] E-mail: taskaev@inp.nsk.su

range of these particles in water or in the body tissues – 5.2 and 7.5 μm, respectively (the size of typical mammalian cell). The stopping power of γ-quantum is significantly lower – 0.3 keV μm^{-1}. Therefore, the main part of the energy release in the nuclear reaction $^{10}B(n,\alpha)^{7}Li$, namely 84%, occurs within a single cell. Therefore, selective accumulation of boron-10 in the tumor cells and subsequent irradiation with neutrons should lead to the destruction of tumor cells with relatively little damage to the surrounding normal cells.

In accordance with the introductory article by W. Sauerwein in a book on the neutron capture therapy [1, p. 3], one can discriminate four periods in the development of the BNCT technique: i) early clinical trials conducted in the United States from 1951 to 1961; ii) pioneering works of Prof. Hatanaka and co-workers, held in Japan from 1968 to the end of the 1980s; iii) clinical trials of deep intracerebral tumors using epithermal neutron beams from nuclear reactors; iv) the use of particle accelerators to produce the beams of epithermal neutrons.

The main requirement for the therapeutic beam of neutrons is often the following: the flux of epithermal neutrons has to be higher than 10^9 cm^{-2} s^{-1}, so that the duration of therapy would be shorter than an hour. Epithermal neutrons are those having the energies in the range from 0.5 to 10 keV. Over the last decade a significant progress has been achieved in development of the accelerating concept of BNCT, particularly in optimization of beam shaping assembly, and there is a better understanding of what is required. It became clear that the term "epithermal neutrons" needs to be clarified. There is a too wide range of neutron energies, so that they all could not be optimized for BNCT. Ideal for BNCT are the neutrons with energies from 1 to 30 keV [1, p. 43 and Fig. 4 at p. 65].

NEUTRON REACTIONS

In clinical trials of BNCT, nuclear reactors are used as the neutron sources. In these reactors, the reaction of ^{235}U fission is used and induced by thermal neutrons. The energy spectrum of neutrons emitted is well described by the distribution $F(E) = 0.77\ E^{½} \exp(-0.775\ E)$ [1, p. 43]. The spectrum extends to 10 MeV, and has a mean energy of around 2 MeV. During BNCT, neutron moderation is performed using a beam shaping assembly comprising the moderator, reflector, absorber, and filters.

For obtaining neutrons by means of the beams of charged particles, nuclear reactions involving the nuclei with loosely bound neutrons are commonly used. There are two types of reactions from the energetic point of view. Exothermic reactions require a minimum kinetic energy of particles. A typical example is the d-d reaction, as a result of which the neutron and tritium are formed. The energy yield of this reaction is 3.265 MeV. This means that the neutron formed has an energy of 2.451 MeV when the energy of deuteron is neglected and a higher energy if this energy is taken into account. One can see that the neutron energy is even greater than the mean energy of neutron in the nuclear fission reaction. Another type of reaction is called endothermic and it requires minimal threshold energy of particles. Near the threshold the neutron energy is very small, so that the use of neutrons for BNCT is very effective.

The main reactions of the neutron generation and graphs of their cross sections are given in Ref. 3. The main parameters of these reactions are presented in Table 1 [4, 5].

In [4] it has been noted that the most attention is being focused on the following four reactions: $^{7}Li(p,n)^{7}Be$, $^{9}Be(p,n)^{9}B$, $^{9}Be(d,n)^{10}B$, and $^{12}C(d,n)^{14}N$. The best of these reactions was recognized as the reaction $^{7}Li(p,n)^{7}Be$ due to the maximum yield and minimum energy of neutrons. However, creating lithium target seemed to be problematic due to a low melting temperature, low thermal conductivity, high chemical reactivity of lithium, and because of production of ^{7}Be radioactive isotope. After 10 years, A. Kreiner in his review on accelerator neutron sources emphasizes [1] that only three reactions providing the generation of neutrons with energies lower than that obtained in the nuclear reactors should be considered. This concerns the threshold reactions $^{7}Li(p,n)^{7}Be$, $^{9}Be(p,n)^{9}B$, and $^{12}C(d,n)^{13}N$. The last of these reactions is actually not considered due to the low yield of neutrons.

The energy of the generated neutrons is in any case higher than the one required for therapy, and therefore neutron moderating is required as a result of elastic and inelastic scattering on the nuclei

of the moderator material. This process is probabilistic. The motion of a neutron is similar to the Brownian particle motion. For this reason, the lower the initial neutron energy, the narrower the spectrum of epithermal neutrons, which is required for BNCT.

Table 1. Basic parameters of the neutron producing reactions

Reactions	Threshold energy, MeV	Particle energy, MeV	Yield, n / mA s	Neutron energy, keV	
				max	min
$^{7}Li(p,n)^{7}Be$	1.880	1.880 1.890 2.500 2.800	0 $6.3\ 10^{9}$ $9.3\ 10^{11}$ $1.4\ 10^{12}$	30 67 787 1100	30 0.2 60 395
$^{9}Be(p,n)^{9}B$	2.057	2.057 2.500 4.000	0 $3.9\ 10^{10}$ $1\ 10^{12}$	20 573 2120	20 193
$^{9}Be(d,n)^{10}B$	0	0 1.500	0 $3.3\ 10^{11}$	3962 4279	3962 3874
$^{13}C(d,n)^{14}N$	0	0 1.500	0 $1.9\ 10^{11}$	4974 6772	4974 5616
$^{12}C(d,n)^{13}N$	0.327	0.327 1.500	0 $6\ 10^{10}$	4 1188	4 707
$d(d,n)^{3}He$	0	0 0.120 0.200	0 $3\ 10^{8}$ $1.1\ 10^{9}$	2451 2898 3054	2451 2123 2047
$t(d,n)^{4}He$	0	0 0.150	0 $4.5\ 10^{10}$	14050 14961	14050 13305

ACCELERATOR BASED EPITHERMAL NEUTRON SOURCES IN THE WORLD

Over the past 30 years, many projects of accelerator neutron sources have been proposed, but only a small number of them are close to successful completion.

As the most advanced the project of Sumitomo Heavy Industries, Ltd. can be considered. A source of neutrons based on a 30-MeV 1 mA cyclotron and a beryllium target was constructed and launched for the Research Reactor Institute of Kyoto University at the site in Kumatori. At this source, since 2012, clinical trials of the technique have been conducted, the results of which have not yet been disclosed because these tests were necessary to obtain a license. Currently, they are building a similar source for the Southern Tohoku General Hospital (Fukushima, Japan) at the expense of the Great East Japan Earthquake Recovery Project.

Currently, the University of Tsukuba in partnership with Mitsubishi Heavy Industry Co., Japan Atomic Energy Research Institute and scientific organization KEK is constructing an 8 MeV 5 mA linac with the beryllium target. To date, a proton beam with a current of 2 mA has been obtained.

A potentially attractive project is being developed by the Cancer Intelligence Care Systems for the National Cancer Center in Tokyo. The company AccSys Technology, Inc. (California, USA – subsidiary of Hitachi) manufactured of 2.5 MeV linac for current of 20 mA. The target with thin layer of lithium on palladium substrate is used for the neutron generation. To date, a proton beam with a current of 11 mA has been obtained.

All three Japanese BNCT centers plan to begin treatment of melanoma in the near future.

Neutron Therapeutics (USA) (previously GT Advanced Technology, and before Twin Greeks Technologies) started construction of 2.6 MeV 30 mA Hyperion™, an electrostatic accelerator of direct action for the clinic of the University of Helsinki (Finland). To generate neutrons, a rotating lithium target will be used.

The fifth successful project should be a project of Budker Institute of Nuclear Physics, which will be discussed in more detail in the next chapter. Currently, a similar facility is being built for a clinic in China: a tandem accelerator with vacuum insulation with a lithium target.

ACCELERATOR BASED EPITHERMAL NEUTRON SOURCE
OF BUDKER INSTITUTE OF NUCLEAR PHYSICS

In 1998, a project of accelerator neutron source for BNCT was proposed, having three main ideas [6]. The first one was aimed at manufacturing a new type of tandem accelerator with vacuum insulation for obtaining a high current proton beam. The second idea concerns the choice of reaction of neutron generation. Despite a low melting point, low thermal conductivity, and high reactivity of lithium, the ^{7}Li(p,n)^{7}Be reaction should be used, since it is most efficient to produce epithermal neutrons. The third idea was in trying to apply a near-threshold neutron generation mode for therapy. In this case, because of the kinematic collimation, the generated neutrons are emitted predominantly forward and have a relatively low energy of about 40 keV.

The first two ideas fully justified themselves - in a compact accelerator a proton beam with an energy of up to 2.3 MeV and a current of up to 7 mA was obtained. The third idea was slightly transformed - it turned out that the most optimal proton energy is not absolutely near-threshold, but energy about 2.3 MeV [7].

Photo of the tandem accelerator with vacuum insulation and its schematic diagram are shown in Fig. 2. A beam of negative hydrogen ions with 23-keV energy from source *1* is rotated by 15° in a magnetic field, focused by magnetic lenses *2*, injected into accelerator *3*, and accelerated to 1-MeV energy. Gas stripping target *7* arranged inside high voltage electrode *6* converts the accelerated negative hydrogen ions into protons, which are accelerated by the same 1-MeV potential difference up to 2 MeV. The potential to high-voltage electrode *6* and five auxiliary accelerating electrodes *5* is supplied from high-voltage power source *9* (sectioned rectifier, not completely depicted) via feedthrough insulator *8* with an ohmic divider. The gas is pumped by turbomolecular pumps *10* (situated at the ion source and accelerator output) and by cryogenic pump *4* through high-voltage electrode louvers.

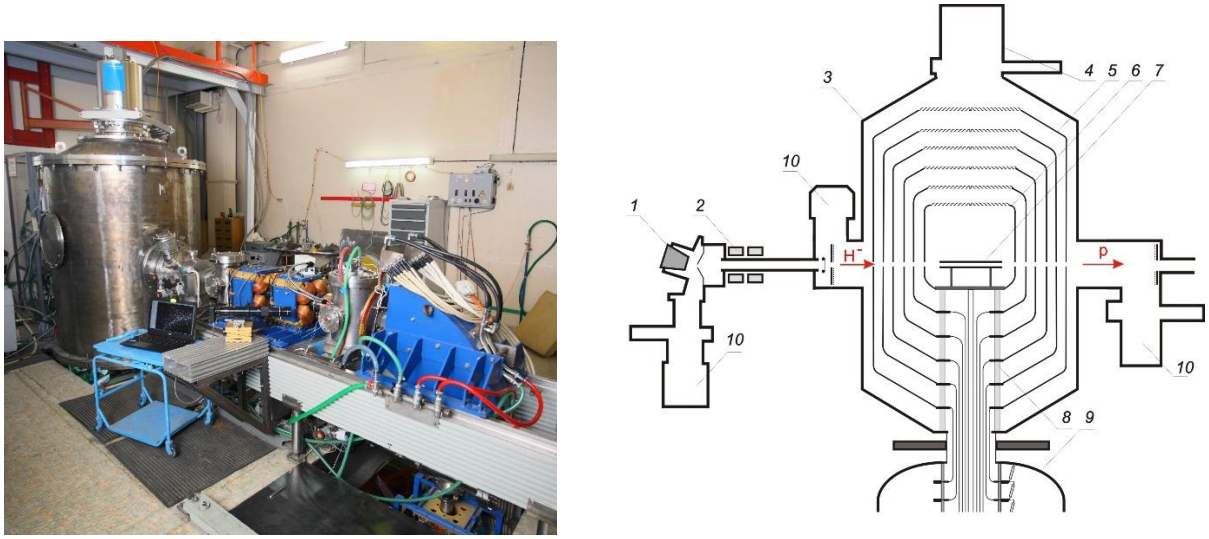

Fig. 2. Tandem accelerator with vacuum insulation: photo (left), scheme (right): *1* – H^- source, *2* – magnetic lenses, *3* – accelerator, *4* – cryogenic pump, *5* – intermediate electrodes, *6* – high-voltage electrode, *7* –gas stripper, *8* – feedthrough insulator, *9* – high-voltage power supply, 10 – turbomolecular pumps. The directions of negative hydrogen ion (H^-) and proton (p) beams are shown by the arrows

The proton beam emerging from the accelerator is rotated downward by the bending magnet and is unfolded along a neutron-generating target with a diameter of 10 cm by the scanner. To generate neutrons, a stationary target is used that is difficult to implement, but provides the optimal quality of the therapeutic neutron beam. In its manufacture the following factors were taken into account 1) Neutron-generating lithium layer should be thin enough for the protons slowed down to the threshold of neutron generation. This will significantly reduce the associated flow of 0.478 MeV γ-quanta and reduce the surface temperature of lithium. 2) Neutron-generating lithium layer should be made of pure lithium for maximum neutron yield. The neutron yield from hydride, oxide, and

lithium fluoride is less than that of pure lithium by 1.43, 2, and 3.3 times respectively. 3) Neutron-generating lithium layer should be in a solid state to prevent the spread of lithium vapor and radioactive isotope beryllium-7 inside the setup. 4) The substrate onto which the lithium neutron-generating layer is deposited should be thin. This will allow researchers to put the optimum moderator close to the place of neutron production and create a therapeutic neutron beam of better quality. 5) The substrate must be cooled intensively to maintain the layer of lithium in the solid state when it is heated by a powerful proton beam. 6) The substrate must be resistant to radiation damage. 7) The substrate should be simple to manufacture. 8) The substrate should be easily removable for its utilization after activation.

The detailed description of the accelerator and the target, as well as the results of the scientific research carried out at that time, is presented in [3]. Below we present the results of the research conducted over the last 4 years.

One of the main results is that the current of the proton beam has been significantly increased: from 1.6 to 7 mA. This was mainly achieved by suppressing secondary charged particles in the accelerator gaps by modernizing the accelerator [8]. An important aspect in increasing the current and improving the stability of the accelerator was the results of the study when the effect of space charge and spherical aberration of magnetic lenses on the phase portrait of the ion beam injected into the accelerator was found [9].

Two important studies have been carried out to develop a neutron-generating target.

Firstly, samples manufactured from lithium, graphite, magnesium fluoride, barium fluoride, aluminum, silicon, titanium, vanadium, stainless steel, copper, molybdenum, and tantalum have been exposed to a proton beam at a tandem accelerator with vacuum insulation. The x-ray and gamma-radiation dose rates and spectra and the neutron-emission dose rate upon the absorption of 2-MeV protons in various materials have been measured along with the residual-activity radiation spectrum [10]. We have found that the absorption of 2-MeV protons in molybdenum or tantalum proceeds at a minimum dose-rate level of accompanying x-ray and gamma radiation and does not lead to the production of fast neutrons and to a residual activity. It is revealed that in order to decrease the undesirable accompanying gamma radiation the substrate of a neutron generating target with a thin lithium layer must be made of molybdenum or tantalum.

Secondly, with the use of a CCD-camera and a remote microscope Infinity K2, the *in situ* observation of blistering of samples prepared from copper and tantalum was performed during their irradiation with a 2-MeV proton beam [11]. The sample temperature during irradiation was measured by a thermistor, pyrometer and an infrared camera. The surface of the irradiated samples was investigated by means of an X-ray diffractometer, laser and electron microscopes.

It is found that the blistering threshold of the copper surface depends on the copper purity. The purer the copper, the higher the threshold is. At a sample temperature of 150°C, the maximum threshold is 3×10^{19} cm^{-2}; the minimum value is seven times lower. Once blisters appear on the copper surface, further irradiation does not cause any more surface modification, which can be due to the formation of holes and cracks when blisters emerge. It is not obvious that after the appearance of blisters on the surface of the copper substrate the target cannot continue to be used to generate neutrons. This is because accumulated hydrogen can escape through holes and cracks formed when blisters emerge. Furthermore, a decrease in the thermal conductivity caused by blisters will not be critical for lithium melting.

It is defined that the samples prepared by four different techniques of depositing tantalum on copper (explosion and diffusion welding, soldering and plasma arc deposition of tantalum and copper powders) are mechanically resistant to both stationary and pulsed thermal loads up to 1 kW/cm^2. Tantalum is much more resistant to blistering than copper. The blistering threshold at 160 to 200°C exceeds 6.7×10^{20} cm^{-2}. At a proton fluence of 3.6×10^{20} cm^{-2}, tantalum surface modification is observed in the form a relief (grid) with a cell size of about 1 μm. It is established that during tantalum irradiation, the sample surface temperature increases, which may be due to a decrease in the thermal conductivity because of the formation of cavities inside tantalum and hydrogen incorporation into the tantalum crystal structure.

The results of these studies led to the fact that in March 2018, after 10 years of use, the target, the construction of which was described in [12], was replaced by a new one, made in the form of a set of thin tantalum tubes [13]. Photos of the targets are shown in Fig. 3.

Fig. 3. Neutron producing target: target used in 2008-2018 (left), new target (right)

A neutron-beam-shaping assembly consisting of a moderator, a reflector, and an absorber is used at accelerator neutron sources to obtain a therapeutic neutron beam for BNCT. For the first time, we have proposed here employing a composite moderator formed by magnesium fluoride near the neutron producing target and aluminum fluoride near the output along with a composite reflector from graphite in the forward hemisphere and lead in the backward hemisphere and generating neutrons via the reaction ^{7}Li(p,n)^{7}Be induced by 2.3-MeV proton beam. By means of a numerical simulation of neutron and gamma-radiation transport, we have shown that the proposed solutions make it possible to shape a therapeutic neutron beam meeting to a great extent the requirements of boron neutron-capture therapy [7, 14].

To confirm the required quality of the neutron beam, together with the colleagues from the University of Tsukuba, irradiation of cell cultures incubated in the medium with boron was carried out. The results turned out to be quite expected [15]: the higher the boron concentration, the better the cells are killed by irradiating neutrons; the higher the doze, the better the cells are killed by irradiating neutrons (Fig. 4).

Even more illustrative result was obtained in experiments on the irradiation of mice with an grafted tumor. The experiments were carried out jointly with the Institute of Cytology and Genetics. In the experiment, 3 groups of SCID mice were used with orthotopic transplantation of U87MG cells (human glioblastoma). The first group of animals was anesthetized four hours after the injection of boronophenylalanine and placed in a thermostable container under a neutron beam for 60 min. The second group of animals was injected with boronophenylalanine, but not irradiated. As intact control, animals were used in which the tumor was not subjected to any effect. To all animals, intracranial transplantation of U87MG cells was carried out on the same day - 32 days prior to the experiment. To do this, a skin incision of 3-4 mm in the region of the bregma was made on the head in the caudal-cranial direction and 5 μl of a suspension containing 0.5 million U87MG cells was injected through the opening in the skull. On the day of irradiation, the volumes of tumors in all the animals were measured in BioSpec 117/16 USR tomograph (Bruker, Germany). Experimental groups were compiled in such a way that the mean values did not differ significantly (mean tumor volumes were 25 ± 7 μl). The results of the experiment are shown in Fig. 5. All mice that were not exposed to radiation died soon enough. Of the 5 mice that were irradiated, three recovered. The tumor volume for them decreased on the 4th day after irradiation, and on the 7th day the tumor was not detected. These mice were euthanized on the 92nd day after tumor transplantation with no signs of disease. Those two mice that were irradiated, but did not recover, had the largest volumes of tumor in the group. The recovery of mice with glioblastoma can be considered to be a very successful experiment for the reason that because of the small size of a mouse it was necessary to irradiate it whole, while in humans the area of irradiation can be localized. As was found using an atomic emission

spectrometer, boron accumulates not only in the tumor, but also in other organs of the mouse (Table 2), and these organs receive a noticeable dose.

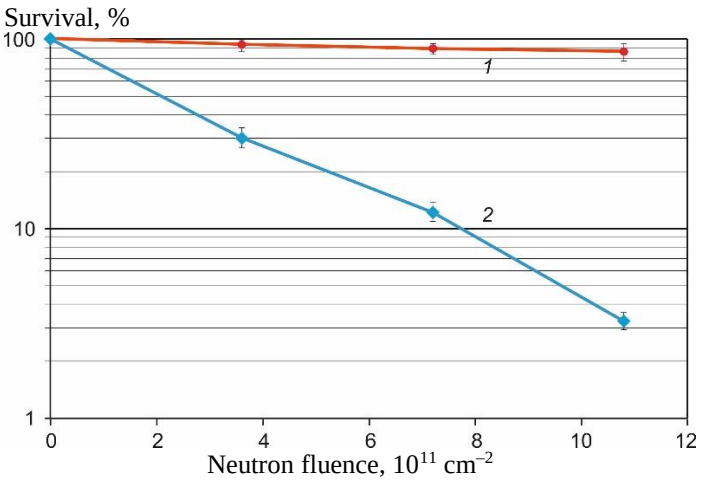

Fig. 4. U251MG cell survival ratio without boron (*1*) and with a boron concentration of 40 ppm (*2*) depending on neutron fluence

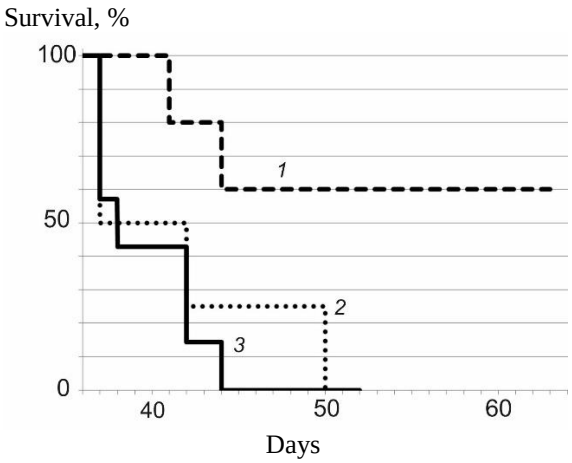

Fig. 5. Kaplan-Meier plot: *1* - group of irradiated mice, *2*- group of mice with boronophenylalanine introduced, but not irradiated, *3* - intact control

Table 2. Concentration of boron (μg/g body weight)

Time after injection, h	In glioma	In the brain	In blood	In the liver	In the kidney	In the skin
1	24	5	30	14	86	34
2.5	20	6	14	5	28	17
5	12	3	4	2	8	5

At present, scientific research is being carried out to obtain a 10 mA 2.3 MeV proton beam and the neutron beam shaping assembly is mounted. Soon it is planned to form a therapeutic neutron beam, suitable for treating patients.

CONCLUSION

A promising method of treatment of many malignant tumors, especially incurable brain tumors, is the boron neutron capture therapy, which is extremely attractive due to selective action directly on tumor cells. It is expected that soon a great number of accelerator sources of epithermal neutrons for BNCT will be created for the widespread introduction of this technique in clinical practice. One such source could be an original accelerator-based source of epithermal neutrons, created in Budker Institute of Nuclear Physics, to which a special attention was attracted in this work. A new type of particle accelerator – the electrostatic tandem accelerator with vacuum insulation – is characterized by high rate of acceleration of charged particles. A stationary proton beam with an energy of 2.3 MeV and a current of 7 mA is obtained. A neutron_generating target, optimal for the formation of epithermal neutron flux that satisfies the requirements of boron neutron capture therapy, was developed and experimentally investigated. A beam of epithermal neutrons is obtained at the facility and scientific research is carried out. The effect of neutron radiation on cell cultures and laboratory animals has been studied. A therapeutic neutron beam suitable for treating patients is planned to be obtained shortly.

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